

# PATENT SPECIFICATION

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 (72) Inventors GEORGE PORTER and DENNIS BOYD



## (54) SYNTHETIC SMOKING PRODUCT

(71) We, GALLAHER LIMITED, a British company, of 138 York Street, Belfast, Northern Ireland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

It is generally recognised that the difficulties in assessing any health hazards which may be associated with cigarette smoking arise from the variety and complexity of the combustion products in the smoke which is inhaled. Many of the combustion products are difficult if not impossible to isolate and are present in such small quantities that their pharmacological activity cannot be properly determined. Various approaches have been used in the past for controlling the combustion products which are inhaled, either by replacing natural tobacco with a substitute material of known composition, or by means of filter plugs through which the smoke is drawn.

We have now conceived a fundamentally new approach in which a smoking material is composed of a matrix of a simple fuel which has mechanical properties, that is flexibility and self-cohesion, and, when made up into cigarette rod form, hardness and porosity, similar to those of natural tobacco, the fuel being impregnated or otherwise associated with volatile solid or liquid constituents which are capable of distilling or subliming into a smoke stream without chemical change and thus providing smoke to be inhaled upon burning of the fuel.

By a simple fuel is meant a material which burns in atmospheric air to produce preferably known simple combustion products of well understood toxicology. The most useful fuel is carbon which burns to simple gaseous oxides having a well understood chemistry and with no unknown health risks. However the use of carbon in the form of charcoal, as has previously been proposed for use as a fuel in analogous fields is unsatisfactory because the carbon fuel in that form is incapable of being handled on conventional cigarette making machinery and other tobacco handling equipment.

We find that the criteria can be satisfied by a fuel comprising a self coherent mass of combustible flexible fibres each of which has a cross sectional dimension of between 5 and 50 microns and which are made of a carbonaceous material containing at least 80% carbon by weight, which material is the product of the controlled pyrolysis of a cellulose-based fibrous material containing only carbon, hydrogen, and oxygen.

The invention also includes a method of preparing a combustible fuel for smoking by human beings, the method comprising providing a self-coherent mass of flexible cellulose-based fibres containing only carbon, hydrogen and oxygen, and subjecting the mass to a controlled pyrolysis until the fibrous mass contains at least 80% carbon by weight, and the fibres have a cross-sectional dimension between 5 and 50 microns.

The basically fibrous nature of the carbonaceous fuel, of which the fibres may have a cross sectional dimension between 5 micron and 50 micron, contributes to the flexibility and mechanical strength of the fuel and to the ability of the fuel particles to hold together without the need for binders or other adhesive aids. The fibres may be additionally crimped or intertwined to further improve the self-cohesion of the material. A matrix of the fuel can be handled on mechanical devices for cigarette making and can be easily formed into cigarettes with an acceptable pressure drop for smoking.

For instance, we have found that fuels made from coarse fibres or mixtures including coarse fibres such as pyrolysed coir or sisal with cross-sectional dimensions up to 300 microns were brittle and not flexible and did not cohere easily together. During handling and transformation of such material to a cigarette form many of these fibres broke and dust and fine particles were formed.

However, for fine fibres such as pyrolysed cotton or ramie with cross sectional dimensions from 5 to 20 microns, the fibres were flexible and self-coherent and could be transformed into an acceptable cigarette form without significant breakage.

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In order for a fibrous fuel to be acceptable in a tobacco substitute it must have a certain hardness as well as the correct pressure drop for smoking. The hardness of the cigarette is conventionally measured by the depression of the surface under a given mechanically applied pressure.

The hardness of the fibrous fuel can be increased by increasing the density to which the fibres are packed inside the wrapper but this also increases the pressure drop of the cigarette to smoking. The acceptable smoking product must contain the fuel packed to the optimum density to achieve both the correct pressure drop and hardness.

In our experimental work, fine fibres of 5 to 20 microns cross-sectional dimension which were sufficiently flexible for incorporation into cigarette-like products were packed at different densities into cigarettes so that they remained as discrete fibres. When the packing density was high the cigarette had acceptable hardness but an undesirably high pressure drop. When packed at low density the pressure drop was acceptable but the cigarettes were not as hard as might be desired. Consequently we find that the fibres of the fuel are preferably agglomerated into clusters. The individual fibres provide coherency between adjacent clusters but the essential porosity of the fuel is provided by the spaces between the clusters of fibres. By a cluster we mean for example a tow or twisted strand of fibres, either straight or crimped, a felt mat of the fibres, or a shredded paper web of the fibres. The tow or strands may of course be additionally formed into a woven cloth which is cut into small pieces.

For instance pyrolysed hemp fibres with cross-sectional dimensions from 5 to 50 microns were agglomerated in the form of twisted tows with a cross-sectional dimension of 1—2 mm. When made up in the form of cigarettes the fibrous fuel was both flexible and non-brittle and gave both an acceptable pressure drop to smoking and acceptable hardness.

The fibres and clusters of fibres are preferably of such a size that the individual fibres

have a cross sectional dimension of between 5 micron and 100 micron and a length of between 1 mm. and 5 cm.; and the clusters have a cross-sectional dimension of between 0.1 mm. and 5 mm. and a length of between 5 mm. and 5 cm.

In order further to change the hardness of the fibrous fuel without incurring a severe change in pressure drop, or vice versa, the fibres or the agglomerates may be aligned along the axis of the cigarette or smoking product instead of being randomly positioned.

The fuel consists of a carbonaceous material which is the product of controlled pyrolysis of a cellulose-based material and which consists of at least 80%, preferably at least 90% carbon by weight. The cellulose-based material is a polymer with a carbon skeleton and containing only carbon, hydrogen and oxygen. The controlled pyrolysis will in general break down the starting material and the result will then be essentially loose linked carbon chains without any significant quantity of side groups containing oxygen or hydrogen. The combustion products from such a fuel will be essentially oxide of carbon and water which satisfy the criteria for a simple fuel. During the pyrolysis the cellulose-based material will suffer weight reduction but its physical characteristics other than simple shrinkage will be largely unchanged. The starting material should therefore also have a flexible fibrous nature.

It is well known that for pyrolysed organic materials, less volatile materials and degradation products are formed during combustion the higher the temperature to which the organic material has been subjected. For instance, a suitable fuel may be made by the controlled pyrolysis of cotton in an inert atmosphere. On burning the fuel under constant suction conditions in a smoking pipe-like device the smoke composition was measured. The following table shows the composition of this smoke for the fuel pyrolysed at 700°C and 900°C and compares the value with flue-cured tobacco smoked under the same conditions.

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Material	N <sub>2</sub> %	O <sub>2</sub> %	CO <sub>2</sub> %	CO %	Methane %	Other Volatiles %
Cotton pyrolysed to 700°C	67	2	16	13	0.3	0.1
Cotton pyrolysed to 900°C	74	1	22	3	<0.01	0.02
Flue-cured tobacco	63	5	20	5	3	4

5 The major components of the smoke from the carbonaceous fuels are nitrogen, oxygen, and oxides of carbon and it can be seen that the material corresponds to that previously cited as a simple fuel.

10 Examples of other suitable fibrous starting materials for the pyrolysis are high purity cellulose based materials such as cotton, cotton linters e.g. after making up into a paper web and shredding, a bast fibre such as ramie, cellulose acetate, or regenerated cellulose such as viscose, and cuprammonium rayon.

15 The fuel itself must have acceptable combustion properties and these are characterised by the rate of burning and the ignition temperature. These values change not only with changes in the pyrolysis conditions of the organic precursor but also with the packing of the fibres. These can also be altered for a given material by the addition of glow sustainers and glow retarders.

20 The fuel must therefore have a suitable ignition temperature and this should not exceed 800°C.

25 For instance it was found that cotton linters in a certain form pyrolysed to 900°C had an ignition temperature in air of 625°C as measured in a conventional thermo-balance. When the fuel was packed into a cigarette-like form it would not burn acceptably. However, if the material was pyrolysed to 500°C the measured ignition temperature was only 510°C and the fuel would burn acceptably in a cigarette form.

30 To convert the fuel to a tobacco substitute it will have mixed or otherwise impregnated with it the volatile solids or liquids which provide the satisfaction to the smoker. These materials should be stable at the temperature to which they are subjected as a result of the burning of the fuel, that is they should distil or sublime without significant decomposition or other chemical change. In practice the volatile solids or liquids will sublime or distil downstream of the burning zone of

the fuel, being heated by conduction and radiation from the burning zone and by contact with the hot combustion products from the fuel. In this way the volatile solids or liquids will not actually be subjected to a temperature as high as that in the burning zone.

50 The volatile constituents may include pharmacologically or physiologically active agents to give the smoker the stimulation of normal tobacco products or other form of stimulation. Examples of such stimulants are nicotine, caffeine, or other pharmacologically active alkaloids. They may be in salt form in which they can be easily applied and from which they evolve during smoking. The volatile constituents may also include a smoke-producing agent which gives a visual and physical impression of smoke from the product, for example by aerosol formation. The smoke-producing agents must also be toxicologically acceptable. Examples of suitable materials are alkanes incorporating between 8 and 32 carbon atoms; high boiling point alicyclics such as decalin; high boiling point ethers such as diisomylether; polyhydric alcohols such as propylene glycol, glycerol, and 1,3 butylene glycol; or glyceryl esters such as triacetin.

75 Further, the volatile constituents may also include flavouring agents to give an aroma to the smoke. Examples of formates, acetates, propionates, and butyrates of terpinols or high molecular weight alicyclic alcohols, ketones, menthol, vanillin, or appropriate natural tobacco extracts.

80 It may also be necessary to include in the fuel a combustion modifying constituent for example for retarding or sustaining glow, or for ash production.

85 If the new material is to be used in a cigarette, a wrapper will normally be necessary. As it is essential that the smoke composition is fully understood, contributions to the smoke stream by uncontrolled combustion of cellulosic products are to be avoided as

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far as possible. The wrapper may therefore either be of an incombustible nature but still sufficiently sensitive to flake off like normal paper, for example in organic films, or a non-porous carbon mat or paper treated in a manner which would prevent the paper combustion products from entering the main smoke stream, such as by coating the surface next to the rod with an intumescent film.

The invention thus enables us to approach the ideal cigarette which incorporates essentially carbon fuel as an open matrix containing only volatiles of known composition and biological activity which are sublimed or distilled off unchanged, and possibly some refractory inorganic materials.

Some examples of materials in accordance with the invention will now be described.

#### Example 1.

A carbon smoking material is produced from heavy weave cotton fabric which is broken down into individual strands before pyrolysis. A sample of about 4 g. is placed in a metal boat and plunged into the tube of a furnace at 500°C which is swept with 1000 ml/minute N<sub>2</sub>. After 15 minutes the sample is removed from the furnace and any further burning is quenched by placing in a beaker with solid carbon dioxide. About 20% of the original sample remains as carbon fibres having a cross sectional dimension between 5 and 50 microns and containing at least 80% carbon by weight.

When the sample is cool it is cut into 10 mm lengths to give a material suitable for packing into a cigarette. The material itself burns too rapidly for cigarette use and is treated with a glow retardant by saturating in a solution of 0.75% sodium dihydrogen phosphate followed by drying in an oven for 48 hours at 55°C.

0.3 g. of this carbon smoking material is then packed into a cigarette form using a slow burning cigarette paper of the papirosi type and a 15 mm long cellulose acetate filter.

A sample cigarette was smoked in a standard cigarette smoking machine which drew 35 ml puffs during a 2 second period every minute. Chromatographic analysis was made of a 5 ml sample of the volatile phase at the end of the middle puff during a standard smoking test. It was found that the total volatile organic phase was only 4.3% of that found in a similar analysis on a standard cigarette made from flue-cured tobacco. Analysis of a sample of the side-stream vapour was only 1% of that from the side-stream in a standard cigarette made from flue-cured tobacco.

#### Example 2.

Cigarettes were prepared in the same manner as in Example 1 but the carbon cigarette was charged with 5 mg of acetophenone and

5 mg of pure nicotine in 120 µl of glycerol distributed inside the carbon material with a syringe and perforated needle.

When these cigarettes were smoked the glycerol produced an aerosol smoke having an optical smoke density comparable to a plain flue-cured cigarette smoke; the three additives were transferred to smoke main stream with less than 1% decomposition.

#### Example 3.

Cigarettes were prepared in the same manner as in Example 1 but the carbon cigarette was charged with 5 mg of anethole, 15.4 mg of nicotine hydrogen tartrate (equivalent to 5 mg of nicotine) and 120 µl of propane 1.2 diol.

When these cigarettes were smoked the propane 1.2 diol again produced an aerosol smoke having an optical smoke density comparable to a plain flue-cured cigarette smoke; the three additives, flavour, smoke producer and nicotine were transferred to smoke main stream with less than 1% decomposition, the acid portion of the salt being decarboxylated and the residue combusted completely.

#### WHAT WE CLAIM IS:—

1. A method of preparing a combustible fuel for smoking by human beings, the method comprising providing a self-coherent mass of flexible cellulose-based fibres containing only carbon, hydrogen and oxygen, and subjecting the mass to a controlled pyrolysis until the fibrous mass contains at least 80% carbon by weight, and the fibres have a cross sectional dimension between 5 and 50 microns.

2. A method according to claim 1, in which the pyrolysed fibres have a cross sectional dimension of between 5 and 20 microns.

3. A method according to claim 1 or claim 2, in which the pyrolysed fibres have a length of between 1 mm and 5 cm.

4. A method according to any one of claims 1 to 3, in which the fibres are agglomerated into clusters.

5. A method according to claim 4, in which each cluster has a cross sectional dimension of between 0.1 mm and 5 mm and a length of between 5 mm and 5 cm.

6. A method according to any one of the preceding claims, in which the pyrolysed fibrous mass contains at least 90% carbon by weight.

7. A method according to any one of the preceding claims, in which, during the controlled pyrolysis, the fibrous mass suffers a weight reduction of substantially 80%.

8. A method according to any one of the preceding claims, in which the cellulose-based fibres are natural fibres.

9. A method according to any one of claims 1 to 7, in which the cellulose-based fibres are regenerated cellulose fibres.

10. A method according to claim 9, in which the regenerated cellulose is viscose.
11. A method according to any one of the preceding claims, in which the fibres are crimped.
12. A method of preparing a tobacco substitute for smoking by human beings, the method comprising a fuel according to any one of the preceding claims and impregnating the fuel with at least one volatile substance capable of distilling or subliming without chemical change on burning of the fuel.
13. A method according to claim 12, in which the volatile substance comprises a pharmacologically or physiologically active substance.
14. A method according to claim 12 or claim 13, in which the volatile substance comprises a smoke-producing substance.
15. A method according to any one of claims 12 to 14, in which the volatile substance comprise a flavouring agent.
16. A method according to any one of claims 12 to 15, in which the fuel is impregnated with a combustion-modifying constituent.
17. A method according to claim 12, substantially as described in any one of the Examples.
18. A fuel for smoking by human beings, the fuel comprising a self coherent mass of combustible flexible fibres each of which has a cross sectional dimension of between 5 and 50 microns and which are made of a carbonaceous material containing at least 80% carbon by weight, which material is the product of the controlled pyrolysis of a cellulose-based fibrous material containing only carbon, hydrogen, and oxygen.
19. A method according to claim 18, in which the fibres have a cross sectional dimension of between 5 and 20 microns.
20. A fuel according to claim 18 or claim 19, in which the fibres have a length of between 1 mm and 5 cm.
21. A fuel according to any one of claims 18 to 20, in which the fibres are agglomerated into clusters.
22. A fuel according to claim 21, in which each cluster has a cross sectional dimension of between 0.1 mm and 5 mm, and a length of between 5 mm and 5 cm.
23. A fuel according to any one of claims 18 to 22, in which the carbonaceous material contains at least 90% carbon by weight.
24. A fuel according to any one of claims 18 to 23, in which the cellulose-based fibrous material comprises natural fibres.
25. A fuel according to any one of claims 18 to 23, in which the cellulose-based fibrous material comprises regenerated cellulose fibres.
26. A fuel according to claim 25, in which the regenerated cellulose is viscose.
27. A fuel according to any one of claims 18 to 26, in which the fibres are crimped.
28. A tobacco substitute for smoking by human beings, the tobacco substitute comprising a fuel according to any one of claims 18 to 27, impregnated with at least one volatile substance capable of distilling or subliming without chemical change on burning of the fuel.
29. A tobacco substitute according to claim 28, in which the volatile substance comprises a pharmacologically or physiologically active substance.
30. A tobacco substitute according to claim 28 or claim 29, in which the volatile substance comprises a smoke-producing substance.
31. A tobacco substitute according to any one of claims 28 to 30, in which the volatile substance comprises a flavouring agent.
32. A tobacco substitute according to any one of claims 28 to 31, in which the fuel is impregnated with a combustion-modifying constituent.
33. A tobacco substitute according to claim 28, substantially as described in any one of the examples.
34. A cigarette rod comprising a tubular wrapper around a filler which includes a fuel according to any one of claims 18 to 27 or a tobacco substitute according to any one of claims 28 to 33.

For the Applicants,  
GILL, JENNINGS & EVERY,  
Chartered Patent Agents,  
53 to 64 Chancery Lane,  
London WC2A 1HN.